

CLAIMS

1. A lithium-nickel-manganese composite oxide which is a composite oxide comprising Li, Ni, and Mn, the composite oxide having a composition represented by  $\text{Li}_x\text{Ni}_y\text{Mn}_z\text{O}_2$  wherein x is  $1+1/9 \pm (1+1/9)/10$ , y is  $4/9 \pm (4/9)/10$ , and z is  $4/9 \pm (4/9)/10$  and having a crystal structure which belongs to the monoclinic system and has a space group of C12/m1 (No. 12).
2. The lithium-nickel-manganese composite oxide of claim 1, characterized by being represented by the general formula  $\text{Li}[\text{Ni}_{0.5-0.5x}\text{Mn}_{0.5-0.5x}\text{Li}_x]\text{O}_2$  wherein x satisfies  $0.03 \leq x \leq 0.15$ .
3. The lithium-nickel-manganese composite oxide of claim 1, characterized by being represented by the general formula  $\text{Li}[\text{Ni}_{0.5-0.5x}\text{Mn}_{0.5-0.5x}\text{Li}_x]\text{O}_2$  wherein x satisfies  $0.05 \leq x \leq 0.11$ .
4. The lithium-nickel-manganese composite oxide of claims 1 to 3, characterized in that in X-ray powder diffractometry using a Cu-K $\alpha$  ray, the peak intensity ratio  $I_{(002)}/I_{(13-3)}$  between the (002) plane and the (13-3) plane in terms of Miller indexes hkl on the assumption of belonging to the monoclinic system is from 1.35 to 1.95.

5. The lithium-nickel-manganese composite oxide of claim 4, characterized in that  $I_{(002)}/I_{(13-3)}$  is from 1.50 to 1.95.

6. The lithium-nickel-manganese composite oxide of claims 1 to 5, characterized in that the lattice constants on the assumption of belonging to C12/m1 (No. 12) of the monoclinic system are as follows:  $a = (5.00 \pm 0.5) \times n_1$  angstroms,  $b = (8.67 \pm 0.87) \times n_2$  angstroms,  $c = (5.05 \pm 0.51) \times n_3$  angstroms,  $n_1$  to  $n_3$  = integer of 1-5,  $\alpha = \gamma = 90.00^\circ$ , and  $\beta = 109.41 \pm 10.94^\circ$ .

7. The lithium-nickel-manganese composite oxide of claim 6, wherein  $n_1$  to  $n_3$  = 1.

8. The lithium-nickel-manganese composite oxide of claim 6, wherein  $n_1$  = 3 and  $n_2$  and  $n_3$  = 1.

9. The lithium-nickel-manganese composite oxide of claims 1 to 8, characterized in that the proportion of lithium-occupied sites in the layers consisting mainly of lithium (at least either of the 2c sites and the 4h sites) in the C12/m1 structure, as determined by the Rietveld analysis, is 93.5% or higher.

10. The lithium-nickel-manganese composite oxide of claims 1 to 10, characterized in that the crystal unit lattice has atomic fraction coordinates represented by at least one of Table 1 and Table 2 and the range of variation thereof is within  $\pm 10\%$  of the coordinate values.

Table 1 Atomic Fraction Coordinates

Atom	wyck	x	y	z
O	4i	0.2600	0.0000	0.7730
O	4i	0.5940	0.0000	0.7730
O	4i	0.9270	0.0000	0.7730
O	8i	0.0850	0.3210	0.2230
O	8i	0.4180	0.3210	0.2230
O	8i	0.7510	0.3210	0.2230
Li	2b	0.0000	0.5000	0.0000
Li	2c	0.0000	0.0000	0.5000
Li	4i	0.3330	0.0000	0.5000
Li	4h	0.0000	0.3380	0.5000
Li	8i	0.3330	0.3380	0.5000
Mn	4i	0.1670	0.0000	1.0000
Mn	4g	0.0000	0.8330	0.0000
Ni	8i	0.3330	0.8330	0.0000

Table 2 Atomic Fraction Coordinates

Atom	wyck	x	y	z
O	4i	0.7400	0.0000	0.2270
O	4i	0.4080	0.0000	0.2270
O	4i	0.0730	0.0000	0.2270
O	8i	0.0850	0.3210	0.2230
O	8i	0.4180	0.3210	0.2230
O	8i	0.7510	0.3210	0.2230
Li	2b	0.0000	0.5000	0.0000
Li	2c	0.0000	0.0000	0.5000
Li	4i	0.6670	0.0000	0.5000
Li	4h	0.0000	0.6620	0.5000
Li	8i	0.3330	0.3380	0.5000
Mn	4i	0.8330	0.0000	1.0000
Mn	4g	0.0000	0.1670	0.0000
Ni	8i	0.3330	0.8330	0.0000

11. The lithium-nickel-manganese composite oxide of claims 1 to 10, characterized by having a sulfur element content of 1,500 ppm or lower.

12. A process for producing the lithium-nickel-manganese composite oxide of claims 1 to 12, characterized by mixing a nickel-manganese composite oxide of the ilmenite structure with a lithium compound and subsequently burning the mixture in an oxygenic atmosphere at a temperature of from 750°C to 1,200°C.

13. The process for lithium-nickel-manganese composite oxide production of claim 12, characterized by

mixing the nickel-manganese composite oxide of the ilmenite structure with the lithium compound in such a proportion as to result in an Li/(Ni+Mn) atomic ratio of from 1.1 to 1.3 and then burning the mixture in an oxygen-containing atmosphere at a temperature of from 750°C to 1,000°C.

14. A process for producing the lithium-nickel-manganese composite oxide of claims 1 to 11, characterized by comprising a first step in which a carbonic acid salt is added to an aqueous solution containing a nickel salt and a manganese salt to precipitate a carbonate of nickel and manganese, a second step in which a lithium compound is added to and mixed with the carbonate of nickel and manganese, a third step in which the mixture is granulated by spray drying, and a fourth step in which the granulated mixture is burned in an oxygen atmosphere at a temperature of 700°C or higher.

15. The process for lithium-nickel-manganese oxide production of claim 14, characterized in that the nickel salt and the manganese salt are any one of the sulfate, hydrochloride, and nitrate or a mixture thereof, and the lithium compound is any one of lithium carbonate, lithium hydroxide, and lithium nitrate or a mixture thereof.

16. The process for lithium-nickel-manganese

composite oxide production of claim 14, characterized in that the carbonic acid salt to be used in the first step is at least one of sodium hydrogen carbonate and sodium carbonate and is used in an amount of 1.0-1.5 equivalents to the total amount of the nickel and the manganese.

17. The process for lithium-nickel-manganese composite oxide production of claim 14, characterized in that the first step is conducted in an operating pH range of 7-10 and an operating temperature range of 20-100°C.

18. The process for lithium-nickel-manganese composite oxide production of claim 14, characterized in that in the second step, pulverization is conducted simultaneously with the mixing to thereby regulate the average particle diameter of the solid ingredients to 1  $\mu\text{m}$  or smaller.

19. The process for lithium-nickel-manganese composite oxide production of claim 14, characterized in that the granulated mixture has an average diameter of 5-30  $\mu\text{m}$ .

20. The process for lithium-nickel-manganese composite oxide production of claim 14, characterized in that after the fourth step, the burned product is washed

with water to remove impurities.

21. The process for lithium-nickel-manganese composite oxide production of claim 14, characterized in that after the first step, the carbonate of nickel and manganese is taken out by filtration, washed, and then redispersed in water.

22. A positive active material for lithium ion secondary batteries, characterized by comprising the lithium-nickel-manganese composite oxide of claims 1 to 11.

23. A lithium ion secondary battery characterized by employing the positive active material of claim 22.